

## SPECIFICATION

EXAMINER'S

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## COMPLETE SPECIFICATION

## Manufacture of Stable Diazo-Salt Preparations

- We, I. G. FARBENINDUSTRIE AKTIEN-GESELLSCHAFT, a Joint Stock Company organised according to the laws of Germany, of Frankfurt a/Main, do hereby declare the nature of this invention and in what manner the same is to be performed, to be particularly described and ascertained in and by the following statement:—
- 10 This invention relates to the manufacture of stable diazo-salt preparations from certain substituted aromatic monoamines of the benzene series by diluting the diazo-solution which is obtainable by
- 15 diazotising the amine in sulphuric acid of at least 75 per cent. strength, with an organic liquid miscible with or soluble in sulphuric acid, preferably at low temperature, isolating the separated
- 20 diazonium sulphate and mixing the dry or almost dry diazonium sulphate, thus obtained, with such a quantity of a mild alkaline agent as corresponds with the sulphuric acid which may adhere
- 25 mechanically to the diazonium sulphate plus 1 equivalent of sulphuric acid per 1 mol of acid diazonium sulphate, and, if necessary, with at least one suitable adjuvant or diluent or both.
- 30 The certain aromatic amines are mono- and poly-halogen-aminobenzenes, nitro-aminobenzenes, dihalogenoluidines, dihalogen-aminophenol ethers, 1-amino-4-methyl-3-halogenbenzenes,
- 35 1-amino-2-methyl-3-halogenbenzenes, 1-amino-5-chloro-2-alkoxy- or -aryloxybenzenes, 1-amino-2-nitro-4- or -5-halogenbenzenes, 1-amino-4-chloro-3-nitrobenzene, 1-amino-2-chloro-4-nitrobenzene,
- 40 1-amino-2-methyl-4-nitro- and -5-nitrobenzene, as well as 2-amino-4-nitro- or 4-amino-3-nitro-1-phenol ethers.
- As suitable organic liquids may be named, for example, the lower members
- 45 of the aliphatic alcohols and ketones.
- The separated diazonium sulphates may be mixed, for example, with the following mild alkaline agents: sodium carbonate, sodium carbonate, sodium borate or
- 50 sodium acetate, magnesia, zinc hydroxide or calcium hydroxide; as adjuvants or diluents there may be used: partly dehydrated aluminium or magnesium sulphate, anhydrous sodium sulphate or
- 55 metal salts of aryl-sulphonic acids.

[Price 1/-]

According to the process described there may be obtained solid, stable and easily soluble diazo-salt preparations. Their aqueous solutions may be used without any further addition for the production of ice-colours.

The following Examples illustrate the invention, the parts are by weight:—

## EXAMPLE 1.

226 parts of acid sulphate of ortho-chloraniline are introduced, while cooling, into a solution of 130 parts of nitrosyl-sulphuric acid in 180 parts of sulphuric acid of 84 per cent. strength. The whole is stirred until the diazotisation is finished; thereupon the concentrated sulphuric acid diazo-solution thus obtained, is stirred into 690 parts of ethyl-alcohol or *n*-propyl alcohol, while cooling. Immediately the separation of the diazonium sulphate begins and is completed by cooling to  $-15^{\circ}\text{C}$ . The diazonium sulphate is filtered and mixed with 100 parts of anhydrous sodium sulphate and dried at about  $45^{\circ}\text{C}$ . 250 parts of partly dehydrated aluminium sulphate of the composition  $\text{Al}_2(\text{SO}_4)_3 \cdot 9\text{H}_2\text{O}$  and 60 parts of anhydrous sodium carbonate are then mixed with the diazonium sulphate.

## EXAMPLE 2.

162 parts of 2:5-dichloraniline are diazotised with a solution of 133 parts of nitrosyl sulphuric acid in 163 parts of sulphuric acid of 86 per cent. strength. The concentrated sulphuric acid diazo-solution is stirred, while cooling, into 400 parts of methanol or ethanol. The separated diazonium sulphate is filtered at  $-10^{\circ}\text{C}$ . to  $-5^{\circ}\text{C}$ ., mixed with 200 parts of anhydrous sodium sulphate and dried at about  $45^{\circ}\text{C}$ . 240 parts of partly dehydrated aluminium sulphate of the composition mentioned in Example 1, 16 parts of magnesia and 35 parts of anhydrous sodium carbonate are then mixed with the diazonium sulphate.

In a corresponding manner the diazonium sulphates from 2:3:4- or 2:4:5-trichloraniline may be separated and worked up to form diazo-salt preparations.

## EXAMPLE 3.

73 parts of dry sodium nitrite are dissolved in 540 parts of warm sulphuric acid of 90 per cent. strength. After cool-

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ing, 1.38 parts of ortho-nitroaniline are introduced at about 20°C. to 25°C. and the whole is stirred until the diazotization is complete. The diazo-solution is then poured, while cooling, into 1300 parts of methanol and the separation of the diazonium sulphate is completed by cooling to -10°C. to -5°C. The diazonium sulphate is filtered and then mixed in the moist state with a mixture of 150 parts of dry borax of the composition  $\text{Na}_2\text{B}_4\text{O}_7 \cdot 2 \text{H}_2\text{O}$  and 220 parts of partly dehydrated aluminium sulphate of the composition mentioned in Example 1.

Instead of diazotising with a solution of 73 parts of sodium nitrite in 540 parts of sulphuric acid it is also advantageous to use for the diazotising operation a solution of 133 parts of nitrosyl-sulphuric acid in 165 parts of sulphuric acid of 85 per cent. strength.

Instead of methanol there may also be used ethanol, propanol, butanol, acetone or another organic liquid miscible with the sulphuric acid diazo-solution obtainable after complete diazotisation and having a sulphuric acid concentration of about 75 to 80 per cent.

In all cases, for the production of the diazo-salt preparations the quantity of the alkaline agent which has to be added must be calculated upon the total acidity of the filtered moist diazonium sulphate.

In a corresponding manner diazonium sulphates and diazo-salt preparations may be obtained from other halogen- or nitro-substituted aminobenzenes, as well as from other aromatic amines above named.

Having now particularly described and ascertained the nature of our said invention and in what manner the same is to be performed, we declare that what we claim is:—

1. The manufacture of stable diazo-

salt preparations by diazotising a mono-, di- or poly-halogenaminobenzene, a nitroaminobenzene, a dihalogentoluidine, a dihalogenaminophenol-ether, a 1-amino-4-methyl-3-halogenbenzene, a 1-amino-2-methyl-3-halogenbenzene, a 1-amino-5-chloro-2-alkoxy- or -aryloxybenzene, a 1-amino-2-nitro-4- or -5-halogenbenzene, 1-amino-4-chloro-3-nitrobenzene, 1-amino-2-chloro-4-nitrobenzene, 1-amino-2-methyl-4-nitro- or -5-nitrobenzene, as well as a 2-amino-4-nitro- or 4-amino-3-nitro-1-phenol ether in sulphuric acid of at least 75 per cent. strength, separating the diazonium sulphate by diluting the sulphuric acid diazo-solution with an organic liquid miscible with or soluble in sulphuric acid, isolating the separated diazonium sulphate and mixing the dry or almost dry diazonium sulphate with the quantity of a mild alkaline agent which corresponds with the sulphuric acid mechanically adhering to the diazonium sulphate plus one equivalent of sulphuric acid for one mol. of acid diazonium sulphate, and, if necessary, with at least one suitable adjuvant or diluent or both.

2. The manufacture of stable diazo-salt preparations, substantially as described with reference to any one of the Examples herein.

3. The stable diazo-salt preparations when prepared or produced by the process of manufacture particularly described and ascertained herein or by any process which is an obvious chemical equivalent thereof.

Dated this 13th day of July, 1934.

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